

Claims

1) Glass filler material for use in dental composites and dental restorations
5 comprising

- a) 65 – 99.95 mol% silicon dioxide (SiO_2),
- b) 0 - 15 mol% aluminum and/or boron oxide (Al_2O_3 , B_2O_3),
- c) 0 - 30 mol % zirconium and/or titanium and/or hafnium oxide (ZrO_2 ,
10 TiO_2 , HfO_2), Y_2O_3 and/or Sc_2O_3 and/or La_2O_3 and/or CeO_2 and/or other
lanthanide oxides,
- d) 0.05 - 4 mol% alkali metal oxides (Na_2O , Li_2O , K_2O , Rb_2O , Cs_2O),
- e) 0 - 25 mol% earth alkali metal oxides (MgO , CaO , SrO , BaO),

wherein the glass filler particles have an average particle size of 0.1 – 20 μm
and wherein these particles have an inner zone and an outer zone up to 1.5
15 μm and wherein the mean concentration of alkali ions of the outer zone relative
to the mean concentration of alkali ions of the inner zone is 10 % or less and
the alkali ions of the inner zone do not significantly migrate to the outer zone.

2) Glass filler material for use in dental composites and dental restorations
20 comprising

- f) 75 – 96.95 mol% silicon dioxide (SiO_2),
- g) 0 - 10 mol% aluminum and/or boron oxide (Al_2O_3 , B_2O_3),
- h) 3 - 30 mol % zirconium and/or titanium and/or hafnium oxide (ZrO_2 ,
25 TiO_2 , HfO_2), Y_2O_3 and/or Sc_2O_3 and/or La_2O_3 and/or CeO_2 and/or other
lanthanide oxides,
- i) 0.05 - 3 mol% alkali metal oxides (Na_2O , Li_2O , K_2O , Rb_2O , Cs_2O),
- j) 0 - 15 mol% earth alkali metal oxides (MgO , CaO , SrO , BaO),

wherein these particles have an inner zone and an outer zone up to 1.5 μm
and wherein the mean concentration of alkali ions of the outer zone relative to
30 the mean concentration of alkali ions of the inner zone is 10 % or less and the
alkali ions of the inner zone are fixed in the particles by a drying process.

- 3) Glass filler material according to claim 1 or 2 comprising
- f) 75 – 96.95 mol% silicon dioxide (SiO_2),
 - g) 0 - 5 mol% aluminum and/or boron oxide (Al_2O_3 , B_2O_3),
 - h) 3 - 30 mol % zirconium and/or titanium and/or hafnium oxide (ZrO_2 ,
5 TiO_2 , HfO_2), Y_2O_3 and/or Sc_2O_3 and/or La_2O_3 and/or CeO_2 and/or other
lanthanide oxides,
 - i) 0.05 - 2 mol% alkali metal oxides (Na_2O , Li_2O , K_2O , Rb_2O , Cs_2O),
 - j) 0 - 5 mol% earth alkali metal oxides (MgO , CaO , SrO , BaO).
- 10 4) Glass filler material according to one of the claims 1 or 2,
wherein the concentration of e) alkali metal oxides is not over 2 mol%,
preferably not over 1 mol%.
- 5) Glass filler material according to one of the claims 1 to 4,
15 wherein the glass filler particles have an average particle size of 0.5 to 3 μm ,
preferably 0.5 to 1 μm .
- 6) Glass filler material according to one of the claims 1 to 3,
wherein the maximal particle size is up to 100 μm preferably up to 5 μm .
- 20 7) Glass filler material according to one of the claims 1 to 6,
wherein the refractive index n_D of the glass filler material n_D is in the range of
1.49 to 1.55.
- 25 8) A method for producing a glass filler material for use in dental composites
and dental restorations with an average particle size of 0.1 to 20 μm by
- g) melting a composition of 54 – 91 mol% SiO_2 , 0 – 13.6 mol% Al_2O_3
and/or B_2O_3 , 0 – 27.3 mol% ZrO_2 and/or TiO_2 and/or HfO_2 and/or Y_2O_3
and/or Sc_2O_3 and/or La_2O_3 and/or Ce_2O_3 and/or other lanthanide oxides,
30 9 – 20 mol% alkali metal oxides, 0 – 22.7 mol% earth alkali oxides at a
temperature of 1200 – 1800 °C for at least 30 minutes,

- h) crushing the melted glass by transferring into cold water or on metal rollers,
 - i) milling the glass granulate obtained by b) to a mean particle size of d_{50} from 0.1 to 20 μm ,
 - 5 j) dealkalizing the glass powder in excess with a dealkalizing agent,
 - k) removing the dealkalizing agent and washing the glass powder with a polar solvent until the filtrate reacts neutral,
 - l) drying the glass powder at a temperature of 200 to 1100 °C for at least 30 minutes.
- 10
- 9) A method according to claim 8,
wherein the melting temperature is from 1400 to 1700 °C, preferably from 1450 to 1550 °C.
- 15 10) A method according to claim 8 or 9,
wherein the dealkalizing agent is an acidic composition.
- 11) A method according to one of the claims 8 to 10,
wherein the dealkalizing agent is an inorganically or organically acid
20 preferably HCl, HJ, HBr, H₂SO₄, H₃PO₄, HNO₃, HClO₄, CH₃COOH, COOH-COOH, H-COOH, citric acid, tartaric acid or polycarboxylic acid.
- 12) A method according to one of the claims 8 to 11,
wherein the polar solvent consists of water or a mixture of water with other
25 polar solvents, preferably ethanol or acetone.
- 13) A method according to one of the claims 8 to 12,
wherein is dealkalizing is performed at temperatures of 50 to 200 °C.
- 30 14) A method according to one of the claims 8 to 13,
wherein the ratio of the glass powder to the dealkalizing agent is 1:5 to 1:1000, preferably 1:10 and more preferably 1:20.

15) Glass filler material for use in dental composites and dental restorations comprising

- a) 75 – 96.95 mol% silicon dioxide (SiO_2),
- 5 b) 0 - 10 mol% aluminum and/or boron oxide (Al_2O_3 , B_2O_3),
- c) 3 - 30 mol % zirconium and/or titanium and/or hafnium oxide (ZrO_2 , TiO_2 , HfO_2), Y_2O_3 and/or Sc_2O_3 and/or La_2O_3 and/or CeO_2 and/or other lanthanide oxides,
- d) 0.05 - 3 mol% alkali metal oxides (Na_2O , Li_2O , K_2O , Rb_2O , Cs_2O),
- 10 e) 0 - 15 mol% earth alkali metal oxides (MgO , CaO , SrO , BaO),

wherein the particles of the glass filler material are produced by the method of claims 8 to 14.

16) A polymerizable dental material containing:

- 15 a) 3 – 80 wt.% of one or more cationically and/or radically curable monomers,
- b) 3 – 90 wt.% of the glass filler material of claim 1 to 7,
- c) 0 – 90 wt.% of one or more radio-opaque fillers,
- d) 0.01– 25 wt.% of initiators, retarders and/or accelerators,
- 20 e) 0 – 25 wt.% of auxiliary agents

17) A polymerizable dental material according to claim 16, wherein the curable monomer is an epoxide monomer.

25 18) Use of a glass filler material according to one of the claims 1 to 7 , for dental filling materials, dental cements, dental bonding materials, dental restorative materials.